Extension of Sediment Geochronology to Coarse-Grained Sediments

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Award Number: N00014-05-1-0088
http://www.vims.edu/

LONG-TERM GOALS

Knowledge of seabed dynamics in nearshore environments is needed to address problems ranging from to the evolution of coastal geomorphology to the changing nature of seabed acoustical properties. One successful approach, used primarily in fine-grained sedimentary environments, has been the application short-lived particle-reactive radioisotopes to quantify sediment deposition and accumulation rates, and to measure the depth and intensity of physical and biological mixing. Unfortunately, the utility of this approach in coarser sediments typical of coastal environments has been hindered by the low inherent concentration of these isotopes in sandy sediments, and the relatively high lower limit of detection using traditional decay counting techniques. The proposed study would attempt to extend this approach into sandy sedimentary environments by taking advantage of a relatively new measurement technique, high-resolution inductively coupled plasma-mass spectroscopy (ICP-MS).

OBJECTIVES

Recent studies by Kenna (2002) have successfully measured Pu concentrations as low as 0.5 fg g^{-1}, a value roughly two orders of magnitude lower than possible by traditional decay counting techniques. $^{239/240}$Pu is one of the major byproducts of thermonuclear explosions, and hence is a key time marker for the period since 1954, the onset of intensive atmospheric weapons testing. This study would attempt to measure $^{239/240}$Pu concentrations in sediment cores collected from Duck, NC. A transect of core samples with a range of textural characteristics will be obtained from other ongoing studies of this area and analyzed for $^{239/240}$Pu. The major goal will be to determine the limits of this new technology in its application to sediment geochronology of sandy sediments.

APPROACH

Recent advances in high-resolution inductively coupled plasma-mass spectroscopy (HR ICPMS) present an unparalleled opportunity to advance the measurement of environmental levels of bomb Pu isotopes in sediment geochronology. A combination of advances in instrumentation and separation techniques has increased the sensitivity of bomb Pu measurement by two orders of magnitude over that typically achieved by traditional decay counting techniques. In fact, the present technology allows the measurement of Pu levels as low as 0.5 fg g^{-1} (Kenna, 2002). For perspective, a rough equivalent in terms of radioactivity is only a single disintegration per day, a quantity that could never be measured using decay counting. HR ICPMS measurement of bomb Pu, therefore, is likely to replace traditional

Report Documentation Page

Form Approved
OMB No. 0704-0188

1. REPORT DATE	2. REPORT TYPE	3. DATES COVERED
30 SEP 2005		**00-00-2005 to 00-00-2005**

4. TITLE AND SUBTITLE	5a. CONTRACT NUMBER
Extension of Sediment Geochronology to Coarse-Grained Sediments	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER
6. AUTHOR(S)	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) **Virginia Institute of Marine Science,College of William and Mary,Gloucester Pt,VA,23062**	8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)	10. SPONSOR/MONITOR'S ACRONYM(S)
	11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited

13. SUPPLEMENTARY NOTES
code 1 only

14. ABSTRACT

Knowledge of seabed dynamics in nearshore environments is needed to address problems ranging from to the evolution of coastal geomorphology to the changing nature of seabed acoustical properties. One successful approach, used primarily in fine-grained sedimentary environments, has been the application short-lived particle-reactive radioisotopes to quantify sediment deposition and accumulation rates, and to measure the depth and intensity of physical and biological mixing. Unfortunately, the utility of this approach in coarser sediments typical of coastal environments has been hindered by the low inherent concentration of these isotopes in sandy sediments, and the relatively high lower limit of detection using traditional decay counting techniques. The proposed study would attempt to extend this approach into sandy sedimentary environments by taking advantage of a relatively new measurement technique, high-resolution inductively coupled plasma-mass spectroscopy (ICP-MS).

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			
unclassified	**unclassified**	**unclassified**	**Same as Report (SAR)**	**4**	

decay counting techniques in the near future for application to problems in marine sediment geochronology. The dramatically lower limit of detectability achieved using this technique should allow us to extend studies heretofore limited to fine-grained sedimentary environments to the sandy environments of the nearshore and inner continental shelf.

Measurement of Pu isotopes will be made using an approach adapted from that of Kim et al. (2000) and Kenna (2002). Sample material (5-10 g) will be spiked with a ^{242}Pu reference solution to monitor chemical yield. The sample will then be combusted at 550° C to remove organic matter before partial acid leaching with HNO_3. Following filtration, the Pu will be oxidized from Pu(III) to Pu(IV) using a sodium nitrate solution. Separation and purification of the Pu isotopes in solution will be made using TEVA-Spec^{TM} ion exchange resin columns as an actinide-nitrate complex. Eluted samples with Pu will be dried and brought to a final volume of 1 ml in 10% HNO_3 for measurement on the ICP-MS. Using a similar approach, Kenna (2002) was able to achieve chemical yields between 70% and 95%.

WORK COMPLETED

All ordering of supplies and laboratory reagents to set up the new Pu extraction facility at VIMS has been completed. The VIMS radiation safety office has expended considerable effort over the past year in order to add Pu to the institute's radioisotope license. This is needed before a Pu standard can be purchased for this project. A major change in the radioisotope licensing authority for VIMS, from region II to region I, has resulted in an unusual delay in NRC approval. VIMS is required to renew its license on a 5 year renewal period and up until last year had dealt with NRC Region II, Atlanta, GA. Because of their familiarity with the VIMS RSO, adding new isotopes had previously been a relatively straightforward and timely process. The change to Region I, King of Prussia, PA, required an entirely new program/license application for the institute. In addition, for the first time, VIMS has had to provide a financial assurance instrument in accord with the type, quantity and risk involved with the particular type of RAM on hand. This process is now complete and we now expect approval by the NRC in the next few weeks.

RESULTS

As indicated above, all of the preliminary preparation and set up for the Pu extraction line has been completed. The unforeseen complexity in renewal of the VIMS radioisotope license, including the addition of Pu to the license, has delayed the purchase of the necessary Pu radioisotope standard. However, the approval process is nearly complete and we anticipate being able to begin extractions later this Fall.

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